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# A gradient surface produced by combined electroplating and incremental frictional sliding

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**Abstract.** A Cu plate was first electroplated with a Ni layer, with a thickness controlled to be between 1 and 2  $\mu\text{m}$ . The coated surface was then deformed by incremental frictional sliding with liquid nitrogen cooling. The combined treatment led to a multifunctional surface with a gradient in strain, chemical content, microstructure, and hardness. The chemical profile was measured by glow-discharge optical emission spectroscopy, showing diffusion of Ni into the heavily deformed Cu layer to a depth of about 40  $\mu\text{m}$ . The microstructure and hardness were characterized and compared with a similarly processed Cu plate without Ni coating, showing a strong effect of the coated layer on the deformation. The experimental results are followed by an analysis of strengthening mechanisms and a discussion of the applicability of the new technique for increasing the durability and lifetime of components exposed to friction and wear, e.g. in wind turbines.

## 1. Introduction

The surface properties of metallic components are critical in determining their durability and lifetime under various conditions, e.g. friction, wear and corrosion. A number of techniques have been developed to improve the surface properties of metallic materials by modifying their chemical compositions and/or microstructures [1-3]. Electroplating is one example widely used to coat one metal by another, for example in order to protect the component against chemical corrosion [4]. Recently, a new technique, namely incremental frictional sliding (IFS), which is able to produce a gradient nanostructured surface layer in a flat sample, has been developed [5]. The resulting microstructural gradient at the surface is known to be effective in enhancing the strength and lifetime of metallic components.

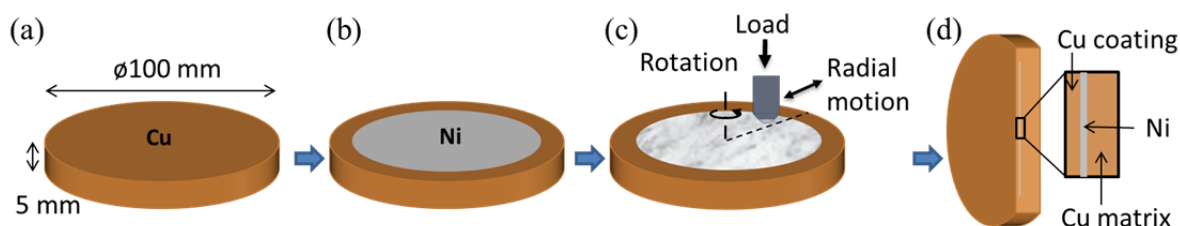
In the present work, we combine electroplating and IFS to produce a multifunctional surface, with a gradient in strain, chemical content, microstructure, and hardness. We choose to coat Ni on a Cu plate since they are completely miscible in the solid state, so a good bonding at the interface is expected. The coated and deformed samples can therefore be used as a model system to investigate the strengthening mechanisms and to evaluate the new combined technique.



## 2. Experimental details

A phosphorus-deoxidized Cu plate (figure 1), 100 mm in diameter and 5 mm in thickness, was ground on one surface and then annealed at 600 °C for 2 h to obtain a fully recrystallized state (Vickers hardness  $46 \pm 3$ ). The main impurities are 0.007Zn-0.024P-0.002Fe-0.002Ni (wt%) as measured by spark optical emission spectroscopy (OES). The central part (80 mm in diameter) of the ground surface was etched for 20 s in a solution containing  $\text{H}_2\text{SO}_4$ ,  $\text{HNO}_3$  and  $\text{HCl}$  to remove the oxide layer and to enhance bonding, during which the edge and the other side of the surface were protected by a Lacomit varnish. Ni was then electrodeposited on the etched surface of the Cu plate. A duration of 6 minutes with a current of 0.5 A at 55 °C (cathode current density 1 A/dm<sup>2</sup>) led to a thickness between 1 and 2 µm (slightly thinner in the center and thicker in the periphery). The water-based solution (pH = 4.5) contained  $\text{NiSO}_4 \cdot 6\text{H}_2\text{O}$  (310 g/L),  $\text{NiCl}_2 \cdot 6\text{H}_2\text{O}$  (45 g/L) and  $\text{H}_3\text{BO}_3$  (37 g/L).

The electrodeposited surface of the Cu plate was then deformed by IFS close to liquid nitrogen temperature for 50 minutes at 200 rpm (figure 1c). To maintain a low temperature, the processing was carried out in 25 interrupted sessions. For each session, the plate was first submerged in liquid nitrogen and then processed for a relatively short duration of 2 minutes without further cooling. During each session, the load (about 100 N) along the normal direction (ND) was moved manually back and forth in the radial direction for about 100 times while the plate was rotating [5]. After deformation a bulging of the entire disk was visible, due to the extension of the processed surface along the radial direction, resulting in a bending of the disk.



**Figure 1.** Schematic of the experimental procedure. (a) Geometry of the Cu plate, (b) electroplating of Ni, (c) deformation by IFS, and (d) coating of protective Cu on the processed surface. The marked and magnified area in (d) indicates the area for microstructural and hardness tests.

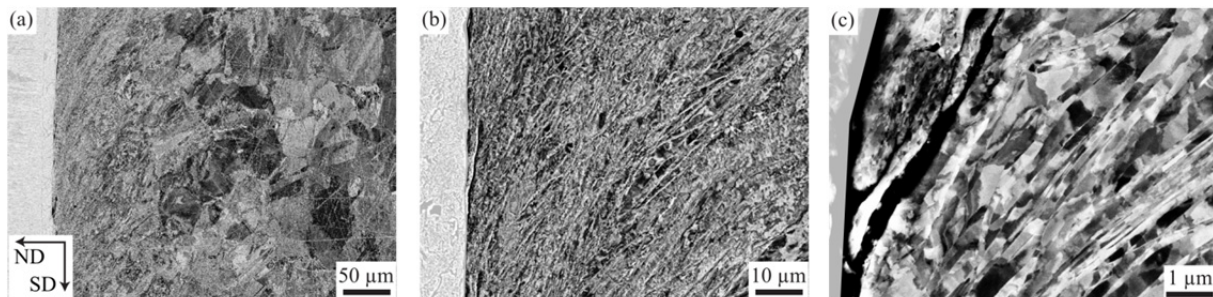
Microstructural observations and hardness tests of the processed plate were carried out on the tangential section (perpendicular to the radial direction) containing the sliding direction (SD) and the ND, at locations between 20 and 35 mm from the center of the plate (figure 1d). For preparation of the tangential section samples, the processed surface was protected by electroplating of Cu, which was done at room temperature for 25 hours (cathode current density 3 A/dm<sup>2</sup>) using a solution containing  $\text{CuSO}_4 \cdot 5\text{H}_2\text{O}$  (72.7 g/L),  $\text{H}_2\text{SO}_4$  (225 g/L) and  $\text{NaCl}$  (98.7 mg/L). Tangential section samples were prepared by mechanical polishing, with a colloidal silica suspension (0.04 µm) used for the final polishing step. The microstructure was investigated using both electron channeling contrast (ECC) and electron backscatter diffraction (EBSD) in a Zeiss Supra 35 scanning electron microscope equipped with a field emission gun. For EBSD analysis, a step size of 50 nm and a cutoff misorientation angle of 2° were used. Vickers hardness tests were carried out on a Struers DuraScan-70, using a load of 245 mN. The Vickers hardness has a high reliability but as the indent diagonal was between 16 and 26 µm in the current case, nanoindentation tests were therefore also carried out in a Hysitron TI 950 TriboIndenter with a maximum load of 2 mN in order to probe the hardness gradient at the top surface.

In order to determine the chemical gradient along the depth, the processed sample surface (without Cu protective coating) was measured by glow-discharge OES in a Horiba GD-Profilier HR.

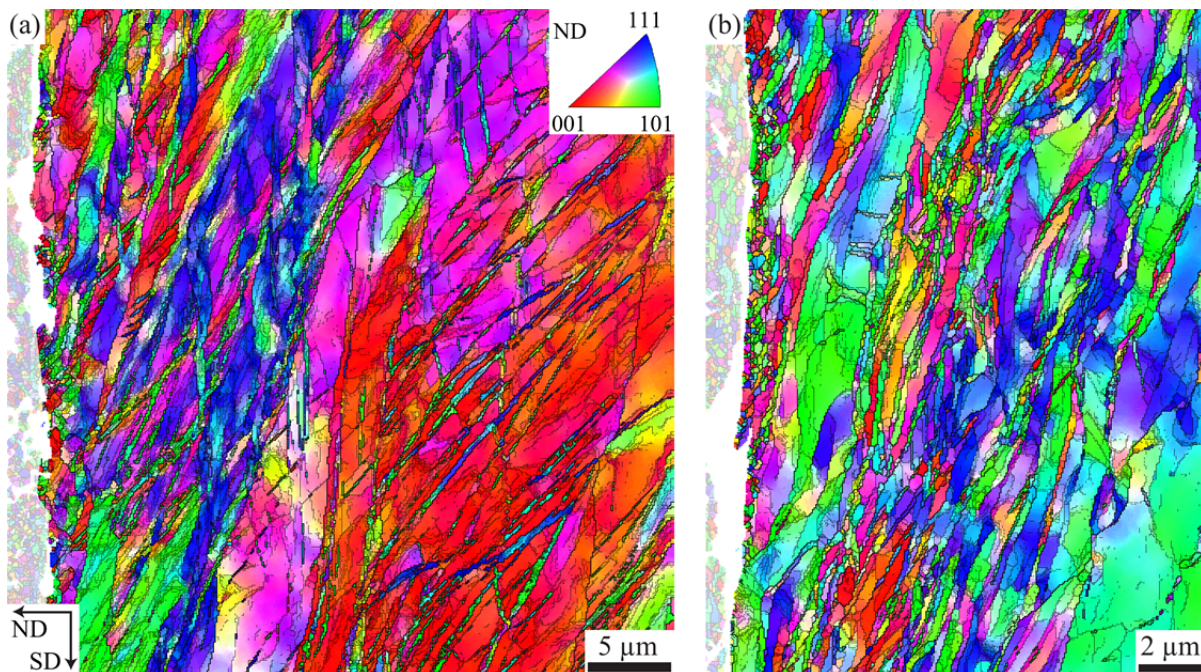


### 3. Results

The combination of electroplating and IFS led to a structural refinement of the processed surface layer. Figure 2 shows examples of ECC observations at the tangential section at different magnifications. The deformation is highly heterogeneous, and the thickness of the heavily deformed zone varies between 5 and 80  $\mu\text{m}$ , below which the initial grain boundaries are discernible. Typically, the top layer of about 40  $\mu\text{m}$  is heavily deformed (figure 2a). This top layer shows a finely spaced lamellar structure, and the extended lamellar boundaries are inclined on average at  $30^\circ$  to the surface, with boundary spacings varying from a few tens of nanometers to a couple of hundred nanometers.



**Figure 2.** ECC observations at the tangential section of the processed sample at different magnifications. The light shaded area in the left part of each micrograph corresponds to the Cu protective coating.

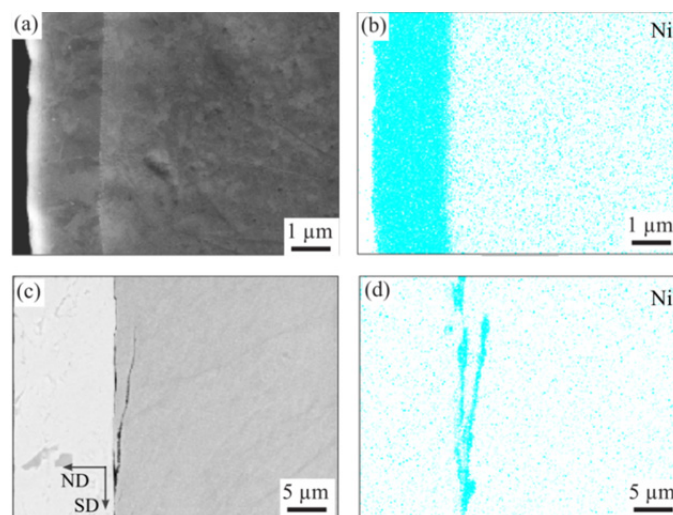


**Figure 3.** EBSD observations at two different locations at the tangential section of the processed surface layer. The colour is coded according to the orientation of the ND, and the Cu protective layer at the surface is shadowed. High angle boundaries ( $>15^\circ$ ) are shown by thick lines and low angle boundaries ( $2-15^\circ$ ) are shown by thin lines.

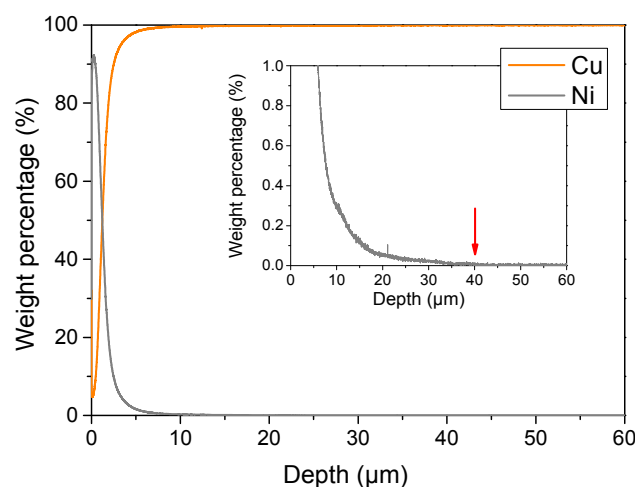
EBSD investigations were carried out in order to study the details of the nanostructured top surface layer, and two examples are shown in figure 3. The fraction of high angle boundaries ( $>15^\circ$ ) was found to be around 50%, and their distribution was highly heterogeneous. The deformation structure

depended strongly on the initial grain orientation. For some grains, well-defined lamellar structures are developed with a high fraction of high angle boundaries, whereas for others, the structure is relatively coarse and contains nanoscale twins. For the top layer of 40  $\mu\text{m}$ , the average boundary spacing measured along the ND in regions of well-defined lamellar structure is about 180 nm. However, the real lamellar boundary spacing is smaller since they are not perpendicular to the ND and many boundaries may be undetected due to the limited spatial and angular resolution of the EBSD technique.

The strain at the topmost layer of 5  $\mu\text{m}$  is highest. It can be seen from figure 2 that the surface has been ploughed and folded. The ploughed volume has a thickness of 3-5  $\mu\text{m}$ , and this ploughing and folding occurred approximately every 30-40  $\mu\text{m}$  along the SD. The ploughing and folding can be best observed from the distribution of the coated Ni layer by energy dispersive X-ray spectroscopy (EDS). Figure 4 shows the morphology of the Ni layer before and after surface deformation. Before deformation, there is a uniform Ni layer on the top of the Cu plate (figure 4a,b); after deformation, the Ni layer is extended and thus overlapped (as a result of the folding) in some places (figure 4c,d).



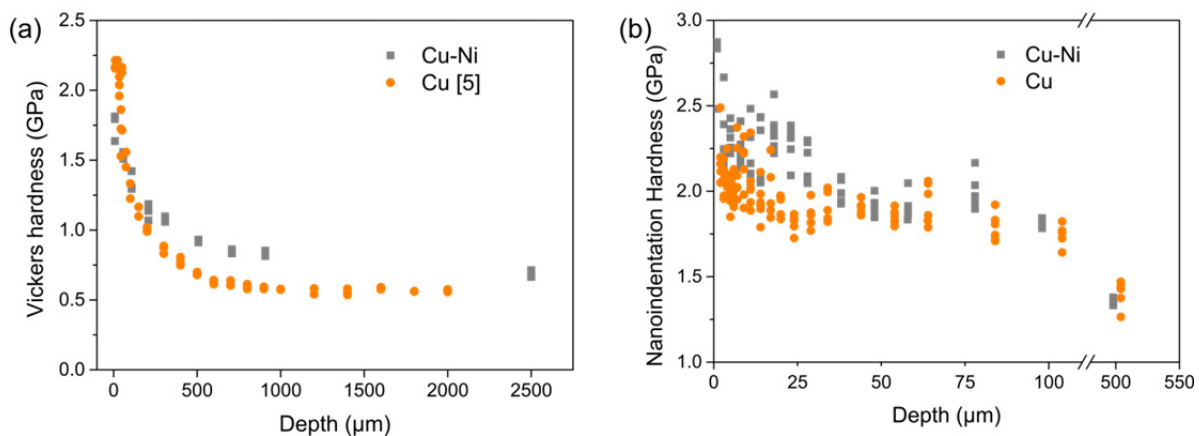
**Figure 4.** Morphology and Ni distribution observed in the tangential section of the surface layer before IFS (a,b) and after IFS (c,d). Note that there is no Cu protective coating in (a), whereas the Cu protective coating in the left part of (c) is seen in light grey. Note the scale bars are different between (a,b) and (c,d).



**Figure 5.** Depth profile revealing diffusion of Ni into the heavily deformed Cu matrix.

EDS is able to differentiate the Ni layer from the Cu matrix, but it cannot accurately detect the amount of Ni diffused into the Cu matrix. The diffusion of Ni was characterized instead by glow-discharge OES, as shown in figure 5. The result shows that on average Ni has diffused to a depth of 40  $\mu\text{m}$  through mechanical alloying, in agreement with the thickness of the heavily deformed surface layer.

The hardness profile of the processed plate has been characterized by Vickers hardness and nanoindentation tests as shown in figure 6 (data in grey), where the Vickers hardness number has been converted to GPa by multiplying 0.0098, to be consistent with a previous study [5]. The hardness decreases gradually from the surface to the bulk interior. Nanoindentation tests resulted in higher hardness values than Vickers hardness tests due to both indentation size effects and different calculation algorithm [6], but the trend is nevertheless similar. The highest hardness ( $2.8 \pm 0.2$  GPa) was recorded 1  $\mu\text{m}$  away from the surface by nanoindentation tests, probably due to the very fine microstructure and a mixture of Ni and Cu. The Vickers hardness at the thickness center ( $0.70 \pm 0.02$  GPa) has also increased from the fully recrystallized state ( $0.45 \pm 0.03$  GPa) as the entire plate was bent during deformation.



**Figure 6.** Vickers hardness (a) and nanoindentation hardness (b) profiles of two IFS processed Cu plates, one with Ni coating (Cu-Ni) and the other without Ni coating (Cu).

#### 4. Discussion

In a previous study [5], a similar Cu plate without Ni coating was deformed by IFS under the same experimental conditions. Without coating of a hard Ni layer, the surface layer (about 100  $\mu\text{m}$  thick) underwent a larger strain, the plastic flow was more parallel to the surface, the deformation was more homogeneous, and the average boundary spacing of the surface layer was smaller. For comparison, the Vickers hardness profile of the previous Cu plate [5] is also shown in figure 6a. The Vickers hardness data are consistent with the microstructural observations. A finer microstructure of the surface layer in the previous Cu plate led to higher hardness of the surface layer in that plate, whereas the bending of the current Cu-Ni plate led to higher hardness of the interior volume compared to that in the Cu plate. In order to further compare the surface layer, nanoindentation tests have also been carried out on the previous Cu plate as shown in figure 6b. It is clear that for the top 40  $\mu\text{m}$ , the hardness of the current Cu-Ni plate is higher than that of the previous Cu plate, which might be related to the heterogeneity of the microstructure in the current Cu-Ni plate as nanoindentation tests were carried out in a very small area.

The results show that the Ni coating has a profound effect on the subsequent surface deformation by IFS. The Ni has a nanocrystalline structure and is much stronger than the fully recrystallized Cu. Therefore during deformation, the Cu matrix is subjected to a very different stress condition with a smaller penetration depth of the steel ball of the loading tool, compared to the situation without Ni



coating. In the current case, compressive deformation was increased while shear deformation was decreased, leading to a much more inhomogeneous microstructure. The enhanced compression of the surface layer resulted in the significant bending of the plate and a high residual stress in the bent plate; whereas the reduced shear deformation led to less structural refinement in the surface layer. Moreover, the plowing occurred at a much smaller length scale (5  $\mu\text{m}$  thick compared to 50  $\mu\text{m}$  in the case without Ni coating), and therefore the processed surface was smoother.

Ni has diffused into the heavily deformed Cu layer to a depth of about 40  $\mu\text{m}$  (see figure 5). The diffusion is assumed to have occurred during deformation, i.e. through mechanical alloying. During deformation at liquid nitrogen temperature, a high concentration of excess vacancies may also be present, which could enhance diffusion rates. More importantly, a high density of dislocations were created and forced to move, leading to long-distance travel of Ni atoms associated with moving dislocations [7]. On the other hand, thermally activated diffusion through the Cu lattice before deformation and through boundaries and dislocations after deformation are negligible during storage at room temperature.

The diffusion of Ni in to the Cu gives rise to a solute strengthening. However, the direct strengthening contribution is typically considered to be small, and therefore the higher hardness measured at the top surface layer in the Ni-coated plate as shown in figure 6b is not likely to be due to solute strengthening. Compared to the Vickers hardness measurements, nanoindentation tests are more sensitive to residual stresses [6], and thus besides heterogeneity, the presence of higher compressive residual stresses in the current Cu-Ni plate may also contribute to the higher nanoindentation hardness measured in that plate.

## 5. Concluding remarks

A combined treatment of electroplating and IFS has been demonstrated in a Cu plate, producing a multifunctional surface layer with a gradient in strain, chemical content, microstructure, and hardness. The hard Ni coating layer influences the deformation behavior of the Cu matrix, and also helps to maintain a smooth surface, which may enhance the friction and wear resistance during subsequent application of the processed material. In order to examine the applicability of the new combined technique, fatigue tests will be carried out as a next step. It has been shown already that gradient structures produced by surface mechanical grinding treatment have a high fatigue resistance due to retardation of both fatigue crack initiation on the hard surface and fatigue crack propagation in the work-hardenable interior [3]. In the current case, a further improvement resulting from the chemical gradient is expected.

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